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PATENT COOPERATION TREATY



PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference B02/0021PC	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP2003/003538	International filing date (day/month/year) 04 April 2003 (04.04.2003)	Priority date (day/month/year) 04 April 2002 (04.04.2002)
International Patent Classification (IPC) or national classification and IPC C07D 487/22		
Applicant BASF AKTIENGESELLSCHAFT		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of <u>4</u> sheets, including this cover sheet. <input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT). These annexes consist of a total of <u>6</u> sheets.
3. This report contains indications relating to the following items: I <input checked="" type="checkbox"/> Basis of the report II <input type="checkbox"/> Priority III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability IV <input type="checkbox"/> Lack of unity of invention V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement VI <input type="checkbox"/> Certain documents cited VII <input type="checkbox"/> Certain defects in the international application VIII <input type="checkbox"/> Certain observations on the international application

Date of submission of the demand 02 October 2003 (02.10.2003)	Date of completion of this report 23 March 2004 (23.03.2004)
Name and mailing address of the IPEA/EP	Authorized officer
Facsimile No.	Telephone No.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/EP2003/003538

I. Basis of the report

1. This report has been drawn on the basis of (Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.):

☐ the international application as originally filed.

☒ the description, pages 1-61, as originally filed,
 pages _____, filed with the demand,
 pages _____, filed with the letter of _____,
 pages _____, filed with the letter of _____.

☒ the claims, Nos. _____, as originally filed,
 Nos. _____, as amended under Article 19,
 Nos. _____, filed with the demand,
 Nos. 1-10, filed with the letter of 27 January 2004 (27.01.2004),
 Nos. _____, filed with the letter of _____.

☐ the drawings, sheets/fig _____, as originally filed,
 sheets/fig _____, filed with the demand,
 sheets/fig _____, filed with the letter of _____,
 sheets/fig _____, filed with the letter of _____.

2. The amendments have resulted in the cancellation of:

☐ the description, pages _____

☐ the claims, Nos. _____

☐ the drawings, sheets/fig _____

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

4. Additional observations, if necessary:

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V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**1. Statement**

Novelty (N)	Claims	1-10	YES
	Claims		NO
Inventive step (IS)	Claims	1-10	YES
	Claims		NO
Industrial applicability (IA)	Claims	1-10	YES
	Claims		NO

2. Citations and explanations

Claimed are cyclic compounds which are derived from benzanelated 5-ring heterocycles, more particularly cyclic quaterbenzoxazoles.

The documents cited in the international search report are already dealt with briefly on page 3 of the application. In connection with **US-A-3 481 945 (D1)**, it is pointed out that the method disclosed therein yields fluoridine, not cyclic tetrabenzimidazole. The applicant is obviously basing this on information which is already found in **US-A-5 180 821 (D2)**. Consequently, D1 is not regarded as relevant and is not taken into consideration in the subsequent procedure.

As regards document D2, the applicant has sufficiently delimited by deleting the use as a phase transfer catalyst. Also endorsed is the argument that, although D2 does disclose unsubstituted cyclic tetrabenzimidazoles, the fields of application mentioned in the present case - light absorbers, OLEDs, pigment dispersers and optical data memories - cannot be inferred from the uses as chelating agents or in electro-optics mentioned in D2.

The substance claim is delimited over D2 by disclaimer and, as regards inventive step, is supported by the novel and non-obvious uses; the requirements of PCT Article 33(2) and (3) appear to be satisfied.

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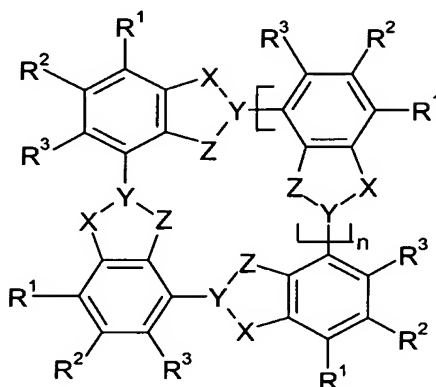
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Finally, as regards the paper by **E. TAUER** in Synthesis, it is assumed that the priority is validly claimed in the present case.

as enclosed to IPER

5 We claim:-

1. The use of cyclic compounds of the formula (I)



where

n is a number in the range from 1 to 7,

X-Y-Z, in each case independently of one another, is O-C=N, N=C-O, NR⁵-C=N, N=C-NR⁵, N⁺R⁵₂-C=N, N=C-N⁺R⁵₂, O-C=N⁺R⁵, N⁺R⁵=C-O, S-C=N⁺R⁵, N⁺R⁵=C-S, S-C=N, N=C-S,

R¹, R² and R³, in each case independently of one another, are H or a substituent from the group consisting of C₁₋₁₂-alkyl, C₁₋₁₂-alkanoyl, C₃₋₇-cycloalkyl, C₆₋₁₂-aryl, C₇₋₁₃-aralkyl, C₇₋₁₃-alkaryl, C₁₋₁₂-alkoxy, C₆₋₁₂-aryloxy, C₁₋₁₂-hydroxyalkyl, a heterocycle, C₆₋₁₂-aroxy, each of which may be substituted, hydroxyl, thiol, halogen, cyano, isocyano, nitro, ammonium, amino, phosphine, phosphine oxide, a sulfonic acid or a derivative thereof, carboxylic acid or a derivative thereof, a derivative of silicon, C₂₋₁₂-alkynyl or C₂₋₁₂-alkenyl, it being possible for the double or triple bonds to be linked directly to the cycloquater skeleton or to be in the chain, a carbamate of the formula

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-NH-CO-OR⁷, a substituted urea of the formula -NR⁷-CO-NR⁷₂, an alkyl carbonate substituent of the formula -O-CO-OR⁷, a sulfinic acid of the formula -SO-OR⁷ or a derivative thereof, a sulfoxide of the formula -SO-R⁷ or a derivative thereof, phosphonic acid or a salt, ester or amide thereof,

it also being possible for R¹ and R² and/or R² and R³, in each case independently of one another, to form unsubstituted or substituted fused ring systems comprising from 1 to 3 rings, which may contain hetero atom groups, or to form unsubstituted or substituted alkylene groups which may be interrupted by hetero atom groups, it also being possible for the fused compounds to be substituted as stated above for the radicals R¹, R² and R³,

it being possible for oxygen atoms in radicals carrying oxygen atoms also to be replaced by sulfur atoms,

it being possible for on average from 0.05 to 100% of the radicals R¹, R² and R³ present in the molecule to differ from hydrogen,

or corresponding heterocyclic compounds in which at least one group -CR¹=, -CR²=, -CR³-CR³= is replaced by -N=,

R⁵, in each case independently of one another, are H, unsubstituted or substituted C₁₋₁₂-alkyl, C₆₋₁₂-aryl, C₇₋₁₃-alkylaryl, unsubstituted or substituted C₁₋₁₂-alkanoyl, unsubstituted or substituted C₇₋₁₃-aryloyl, oligoethylene glycol having 1 to 6 oxygen atoms, oligoethylene glycol ether having 1 to 6 oxygen atoms, imidazoymethyl or a corresponding radical in which a nitrogen atom is substituted by a C₁₋₁₂-alkyl radical and may carry a positive charge and a C-H group in the ring may be replaced by C-(C₁₋₁₂-alkyl), or (1-C₄₋₆-lactam)methyl, which may be C₁₋₁₂-alkyl-substituted on the ring,

R⁷, in each case independently of one another, are H, C₁₋₁₂-alkyl or C₆₋₁₂-aryl,

and tautomeric structures thereof

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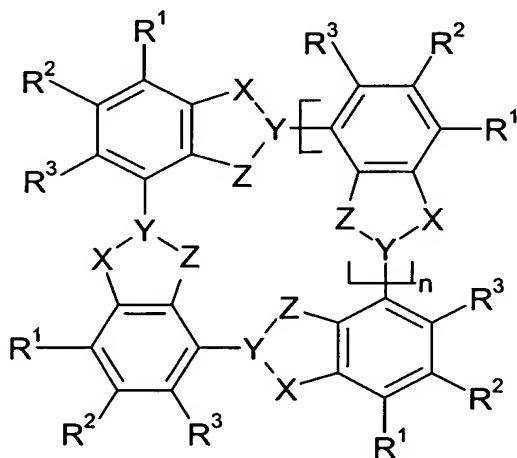
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or metal complexes of the cyclic compounds or complexes of the cyclic compounds with mineral acids,

chloride, sulfate, bisulfate, phosphate, hydrogen phosphate, nitrate, BF_4^- or methanesulfonate being present as opposite ions X^- in the case of cationic cyclic structures,

as light absorbers, materials for hole injection layers in OLEDs, light-emitting compounds in OLED and synergistic agents for the dispersing of pigments or for optical data storage.

2. The use as claimed in claim 1, wherein a cyclic compound of the formula (I)



(I)

where

n is an integer in the range from 1 to 7,

X-Y-Z, in each case independently of one another, is O-C=N, N=C-O, NH-C=N, N=C-NH, S-C=N or N=C-S,

R¹, R² and R³, in each case independently of one another, are H or a substituent from the group consisting of C₁₋₁₂-alkyl, C₁₋₁₂-alkanoyl, C₃₋₇-cycloalkyl, C₆₋₁₂-aryl, C₇₋₁₃-aralkyl, C₇₋₁₃-alkaryl, C₁₋₁₂-alkoxy, C₆₋₁₂-aryloxy, C₁₋₁₂-hydroxyalkyl, a heterocycle, C₆₋₁₂-aroyl, each of

which may be substituted, hydroxyl, thiol, halogen, cyano, isocyano, nitro, ammonium, amino, phosphine, phosphine oxide, a sulfonic acid or a derivative thereof, a carboxylic acid or a derivative thereof or a derivative of silicon,

it also being possible for R^1 and R^2 and/or R^2 and R^3 , in each case independently of one another, to form unsubstituted or substituted fused ring systems comprising from 1 to 3 rings, which may contain hetero atom groups, or to form unsubstituted or substituted alkylene groups which may be interrupted by hetero atom groups,

it being possible on average for from 0.01 to 12 of the radicals R^1 , R^2 and R^3 present in the molecule to differ from hydrogen,

or corresponding heterocyclic compounds in which at least one group $-CR^1=$, $-CR^2=$ or $-CR^3$ is replaced by $-N=$,

or metal complexes of the cyclic compounds,

as light absorbers, materials for hole injection layers in organic light-emitting diodes (OLED)

or as synergistic agents for the dispersing of pigments,

is used.

3. The use as claimed in claim 1 or 2, wherein the light absorber is a UV absorber and/or Vis absorber.

4. The use as claimed in any of claims 1 to 3, wherein the cyclic compound of the formula (I) is used in soluble, partly soluble or insoluble form in an application medium, it also being possible for solid solutions with other colorants to be present in the insoluble form.

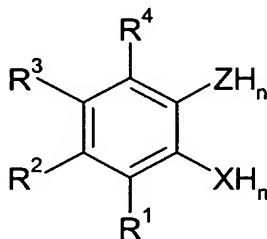
5. The use as claimed in any of claims 1 to 4, wherein R^1 , R^2 , R^3 have the same meanings for all positions.

6. A cyclic compound or a metal complex of the cyclic compounds or a complex of the cyclic compounds with a mineral acid, as defined in claim 1 or 2, with the exception of cyclic compounds where

X-Y-Z is N=C-O, NH-C=N or N=C-NH,

R¹, R² and R³ are H or C₁₋₆-alkyl.

7. A process for the preparation of a cyclic compound of the formula (I) as claimed in claim 6 by cyclization of a compound of the formula (II)



(II)

where

R¹, R², R³, X and Z are as stated,

R⁴ is -COOH or a derivative thereof and

n in each case is 1 or 2, to obtain the stoichiometry,

it also being possible for OH groups to be present as alkali metal salt or ammonium salt groups and/or for NH₂ groups to be present in protonated form or derivative form as -NO, -NO₂, -N=N-aryl, =NOH, =NH, and it being possible for the cyclization to be carried out in the presence of metal salts, metal powders or Lewis acids as templates and in the presence of condensing agents or under dehydrating conditions.

8. A process for the preparation of a complex of a cyclic compound by the preparation of the cyclic compound by a process as claimed in claim 8 in the presence of metal salts or metal powders as templates or by reaction of a cyclic compound as claimed in claim 7 with a metal salt or metal powder.

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9. The use as claimed in any of claims 1 to 5 as a light absorber for coloring high molecular weight organic materials.
- 5 10. A thermoplastic molding material, finish or coating composition comprising a light absorber as defined in any of claims 1 to 5.

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